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A Mechanism For Inducing Climatic Variations
Through Ozone Destruction: Screening of
Galactic Cosmic Rays by Solar and Terrestrial
Magnetic Fields

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ABSTRACT

The evidence that variations in the sun itself are responsible for climate changes is substantial and hence it is frequently suggested that the sun's near-ultraviolet radiation is significantly variable. Whether or not that inference is true, the evidence is strong that the sun's magnetically associated phenomena have long-term variations, aside from the familiar 11-year cycle. A long period of low solar activity and enhanced cosmic-ray production of NO_x in the stratosphere will reduce the ozone abundance.

A perturbation analysis, allowing for temperature and opacity feedbacks, is developed to calculate depletions in the O_3 abundance and reductions of stratospheric solar heating that result from increases in NO_x concentration. A pair of "perturbation coefficients" give the reduction in O_3 and temperature through the stratosphere for a specified NO_x increase. This type of analysis illustrates the tendency for various levels to self-heal when a perturbation occurs.

Physical arguments appear to indicate that the expected sign of the climatic effect is correct, with colder surface temperatures produced by reduced magnetic shielding. In addition, four qualitative reasons are suggested for thinking significant ozone reductions by cosmic-ray influxes will lead to an increased terrestrial albedo from stratospheric condensation. In this view long-term ($\sim 10^4$ years) climatic changes have resulted from secular geomagnetic variations while shorter ($\sim 10^2$ years) excursions are related to changes in solar activity.

1. Introduction

Climatic changes have been attributed to internal exchanges of energy (among atmosphere, oceans, ice caps) as well as to external modulations, and even the relative importance of the two classes of mechanism is not settled (Schneider and Dickinson, 1974). Nevertheless, a scholarly historical study by Eddy (1976) provides convincing evidence that the coldest part of the "little ice age", between about 1650 and 1715, coincided with a near absence of solar activity (the "Maunder minimum").

Schneider and Mass (1975) have obtained a striking agreement between the global surface temperatures since 1600 and temperatures computed with a simple formula based on radiative equilibrium. This approach relates the equilibrium temperature to the solar constant and then connects the solar constant to the sunspot number by the Kondratyev-Nikolsky (1970) empirical formula, which has no theoretical justification but was thought to describe 11-year variations in the solar constant. This sunspot formula implies an increase in solar flux up to 2.5 percent as the Wolf sunspot number increases from 0 to 80 (indicating moderate activity) and then slowly decreases again to 1.9 percent enhancement for Wolf numbers of 200 (high activity). As Schneider and Mass emphasize, it is not important whether the formula gives a correct relation between the solar constant and sunspots; it may merely serve

as a link in a convenient relationship between surface temperature and solar activity. The fact that the link goes through solar flux as the physical connection does not mean it is so. Any effect, such as solar particle emission, solar magnetic fields, or atmospheric transparency (or, as we shall argue below, all three) may just as well be the physical connection between sunspot and climate.

Suess (1965, 1968, 1971, 1974) has shown, by two independent correlations, that climate is associated with solar activity over the past 6000-8000 years. First, production of radioactive ^{14}C (by galactic cosmic rays) fluctuates due to changes in solar magnetic activity. Second, the ^{14}C level seems to fluctuate with climate, in the sense that prolonged high solar activity leads to warm winters.

Long-term changes (of the order of 10^4 years) in the ^{14}C level are likely due to a change in the geomagnetic dipole moment (Ramaty, 1965; Suess, 1965). For still longer periods it seems reasonable to conclude that faunal extinctions that are correlated with reversals of the earth's magnetic field over the past 2.5 million years (Hays, 1971; Opdyke, 1972) are due to climatic changes induced by the increased cosmic-ray deposition in the middle atmosphere (Harrison, 1968), rather than by direct cosmic-ray effects at the earth's surface (Uffen, 1960).

Nevertheless, it has not been clear what physical mechanism would relate cosmic rays to the Earth's climate. In the meantime a rather different line of study has indicated a modulating influence of the sun on the stratosphere, not through variable solar radiation but through the sun's 11-year cycle of solar-wind activity.

Warneck (1972) and Brasseur and Nicolet (1973) noted that cosmic-ray ionization in the stratosphere produced free N and N^+ , which in turn react with oxygen to form NO_x . Although this source of NO_x (which catalytically destroys ozone) is relatively minor, Ruderman and Chamberlain (1975) showed that the observationally known 11-year variation in cosmic-ray ionization over the polar cap implied a predictable 11-year variation of ozone, both over the polar cap and, with some months' time lag, at lower latitudes as well. Analysis of the available data (Angell and Korshover, 1973, 1975) strongly supports the existence of an 11-year ozone cycle with approximately the predicted phase lag from sunspot maximum. The observed amplitude seems to be stronger than was expected from simple ionization and subsequent production of NO_x . Hence there is reason to think additional chemical reactions occur - perhaps involving negative ions - that could be especially important in destroying ozone over the polar caps (Ruderman, Foley, and Chamberlain, 1976).

The combination of the historical relationship between solar activity and climate with a chemical link between low-energy cosmic rays and the ozone balance may be the crucial clue: It suggests that heliomagnetic and geomagnetic control of cosmic-ray fluxes and ozone destruction in the stratosphere is a principal factor influencing climate.

Alternative or additional causal mechanisms are nevertheless possible. Ruderman (1974) has suggested that faunal extinctions could be induced by explosions of nearby supernovae. The dramatically increased x-ray or cosmic radiation at the earth could reduce the protective ozone shield, subjecting life forms (especially DNA) to increased ultraviolet exposure. Life would be especially vulnerable to radiation from giant solar flares during this period.

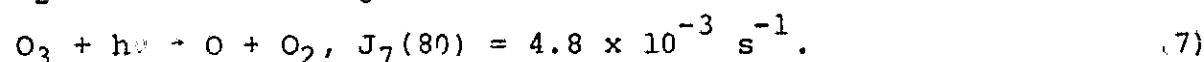
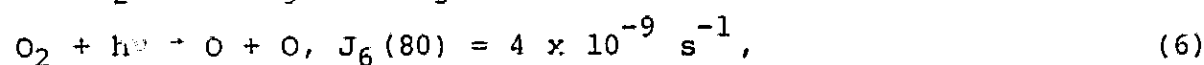
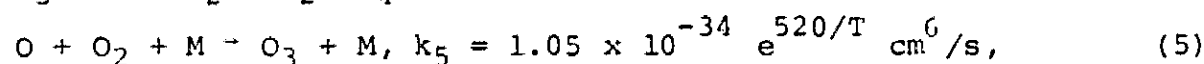
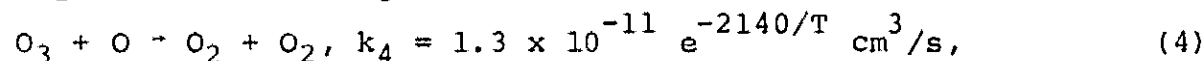
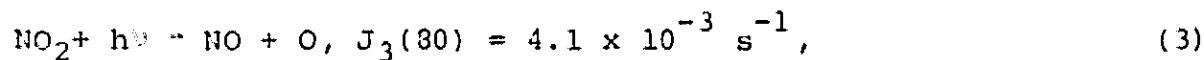
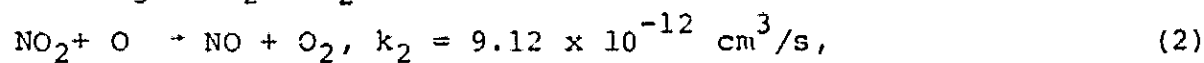
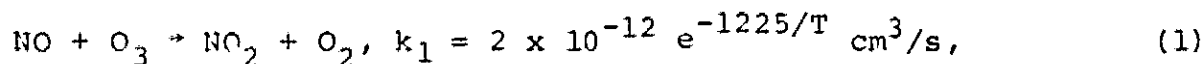
Reid et al. (1976) have proposed that faunal extinctions occur during times of geomagnetic field reversals because the stratosphere is then more susceptible to the occasional solar cosmic rays. As with Ruderman's supernova, the particle bombardment creates stratospheric NO_x , which depletes ozone, making surface life susceptible to solar ultraviolet radiation.

Whether or not life has been directly subjected to hazardous increases in ultraviolet radiation, long-period changes in the heliomagnetic or geomagnetic field seem certain to affect the ozone abundance in the stratosphere. In the next section we develop a perturbation treatment to estimate

the redistribution with height and the magnitude of total ozone decrease accompanying a rise in NO_x . The concluding section examines the likely influence on climate, including the difficulty posed by certain "feedback" mechanisms. We propose that the stratosphere itself is the primary cause of climate changes through increased water condensation.

2. Perturbation Treatment of Stratospheric O_3 Abundance

Consider an oxygen-nitrogen atmosphere. The principal reactions with rate coefficients, k , and dissociation rates at 80 km, $J(80)$, are:



The rates of reaction and photolysis are those quoted by McElroy *et al.* (1974). (Photodissociation (3) of NO_2 is negligible as a source of O atoms.)

In the Earth's stratosphere, reactions involving hydrogen (OH, HO_2 , etc.) are important at the higher altitudes. Also,

HNO_3 is an important component of the total NO_x in the lower stratosphere, but adjustments in relative abundances of NO and NO_2 to the total NO_x can be made as an afterthought.

Given the $[\text{O}]$, $[\text{O}_3]$ concentrations the $[\text{NO}]$ ratio in photochemical equilibrium is

$$r_1 \equiv \frac{[\text{NO}]}{[\text{NO}_2]} = \frac{[\text{O}] k_2 + J_3}{[\text{O}_3] k_1 + [\text{O}] k_2 + J_3} \quad (8)$$

and, similarly, $r_2 = 1 - r_1$. If r_1 is known, the $[\text{O}]$ and $[\text{O}_3]$ concentrations are solutions of quadratic equations.

Define

$$A = k_4 (k_1 [\text{NO}] + 2J_7), \quad (9)$$

$$B = k_2 [\text{NO}_2] (k_1 [\text{NO}] + J_7) + k_1 [\text{NO}] k_5 [\text{O}_2] [\text{M}] + 2k_4 J_6 [\text{O}_2], \quad (10)$$

and

$$C = -2k_5 [\text{O}_2] [\text{M}] J_6 [\text{O}_2]. \quad (11)$$

Then we have

$$[\text{O}_3] = \frac{(B^2 - 4AC)^{1/2} - B}{2A} \quad (12)$$

Similarly, with

$$A' = k_4 (k_2 [\text{NO}_2] + 2 k_5 [\text{O}_2] [\text{M}]) , \quad (13)$$

$$B' = k_2 [\text{NO}_2] (k_1 [\text{NO}] + J_7) \\ + k_1 [\text{NO}] k_5 [\text{O}_2] [\text{M}] - 2 k_4 J_6 [\text{O}_2] , \quad (14)$$

and

$$C' = -(k_1 [\text{NO}] + J_7) 2 [\text{O}_2] J_6 , \quad (15)$$

we obtain

$$[\text{O}] = \frac{(B'^2 - 4 A' C')^{1/2} - B'}{2 A'} \quad (16)$$

Equations (12) and (16) may be solved by iteration with equation (8) to obtain equilibrium abundances of $[\text{NO}]$, $[\text{NO}_2]$, $[\text{O}]$, and $[\text{O}_3]$, when the total $[\text{NO} + \text{NO}_2]$ is specified. We shall call this the "equilibrium solution".

Now let the total NO_x be varied by a relative amount

$$\delta = \frac{\Delta [\text{NO} + \text{NO}_2]}{[\text{NO} + \text{NO}_2]} . \quad (17)$$

We define the intermediate quantities

$$F = \frac{2C}{G^2 - GB} + \frac{1}{A} , \quad (18)$$

where

$$G = (B^2 - 4AC)^{1/2} . \quad (19)$$

Then if the relative change in $[NO]$ and $[NO_2]$ were known we would have

$$\frac{\Delta[O_3]}{[O_3]} = -F \Delta A - \Delta B/G \quad (20)$$

and

$$\frac{\Delta[O]}{[O]} = -F' \Delta A' - \Delta B'/G' - \Delta C' 2A' / (G'^2 - G'B') , \quad (21)$$

where F' , G' are defined analogously to F , G .

In the same way we may write the perturbations $\Delta[NO]$, $\Delta[NO_2]$ in terms of $\Delta[O]$, $\Delta[O_3]$ from equation (8), where

$$\begin{aligned} \frac{\Delta[NO]}{[NO]} &= \delta + \frac{\Delta r_1}{r_1} \\ &= \delta + \frac{r_2}{j} \frac{\Delta[O]}{[O]} - \frac{r_2 \Delta[O_3]}{[O_3]} , \end{aligned} \quad (22)$$

and where

$$j = 1 + J_3/k_2 [O] \quad (23)$$

Similarly we have

$$\frac{\Delta[NO_2]}{[NO_2]} = \delta - \frac{r_1}{j} \frac{\Delta[O]}{[O]} + r_1 \frac{\Delta[O_3]}{[O_3]} \quad (24)$$

Equations (20), (21), (23), and (24) form a complete set of equations that fix the variations of $[O]$, $[O_3]$, $[NO]$, and $[NO_2]$ once δ of equation (17) is specified.

We now define

$$f = \frac{([O_2][M] k_5 + [NO_2] k_2) k_1 [NO]}{G} + k_4 k_1 [NO] F, \quad (25)$$

$$g = \frac{(J_7 + [NO] k_1) k_2 [NO_2]}{G}, \quad (26)$$

$$f' = \frac{([O_2][M] k_5 + [NO_2] k_2) k_1 [NO]}{G'} - \frac{4[O_2] J_6 k_1 [NO] A'}{G'^2 - G'B'}, \quad (27)$$

and

$$g' = \frac{(J_7 + [NO] k_1) k_2 [NO_2]}{G'} + k_4 k_2 [NO_2] F' . \quad (28)$$

The solutions are

$$\frac{\Delta_1 [O_3]}{[O_3]} = - \left[\frac{gf' - fg' + j(f+g)}{r_2 f' - r_1 g' + j(1+r_1 g - r_2 f)} \right] \delta \equiv -\phi_1 \delta , \quad (29)$$

$$\frac{\Delta_1 [O]}{[O]} = - \left[\frac{j(gf' - fg') + j(f' + g')}{r_2 f' - r_1 g' + j(1+r_1 g - r_2 f)} \right] \delta \equiv -\gamma_1 \delta , \quad (30)$$

and

$$\frac{\Delta_1 r_1}{r_1} \equiv - \frac{r_2}{r_1} \frac{\Delta r_2}{r_2} = \left[\frac{r_2 (jf + jg - f' - g')}{r_2 f' - r_1 g' + j(1+r_1 g - r_2 f)} \right] \delta \equiv \rho_1 \delta , \quad (31)$$

where the identities define the perturbation coefficients ϕ_1 , γ_1 , and ρ_1 . Then $\Delta[NO]$ and $\Delta[NO_2]$ are given by (22) and (24).

There are now two important feedback mechanisms to consider. (1) A change in $[O_3]$ and $[NO_2]$ alters the solar

heating, which alters the temperature, which changes the rate coefficients. (2) A change in $[O_3]$ at height z alters the opacity (principally for O_3 photodissociation), which changes the photolysis rate J_7 .

Of course, if the temperature changes, the rate of cooling also changes, but for small perturbations the modified equilibrium temperature may be estimated from Dickinson's (1973) perturbation analysis. Let η_o ($^{\circ}K/day$) be the heating rate per molecule due to O_3 absorption of sunlight, η_x the same for NO_2 , and a (day^{-1}) Dickinson's cooling rate for small temperature changes. The rate of temperature response to departures from equilibrium values is

$$\frac{dT}{dt} = \Delta(\eta_o [O_3]) + \Delta(\eta_x [NO_2]) - a \Delta T. \quad (32)$$

The heating rate η_o suffers from the overhead opacity and can be tabulated as a function of the integrated overhead ozone, I_0 , for a given model. When the perturbed system again reaches a steady state the temperature change is

$$\begin{aligned} \Delta_1 T = & \frac{\eta_x [NO_2] \delta}{a} \left(1 - \frac{r_1}{r_2} c_1\right) \\ & - \frac{\eta_o [O_3]}{a} [\tau_1 \delta + c(I_0) \Delta I_0] , \end{aligned} \quad (33)$$

where

$$\sigma(I_0) = - \frac{dn_0}{n_0 dI_0} \quad (34)$$

is an effective attenuation cross section for the heating sunlight. Since ΔI_0 will be the opposite sign of δ , the term in brackets represents a tendency for self-healing: A positive δ leads to less heating in the upper levels, but more sunlight penetrates to lower levels, where the diminished heating starts to recover. The $[NO_2]$ term also tends to compensate the $[O_3]$ absorption, especially at lower levels, because the radiation it absorbs is essentially unattenuated (Ramanathan, et al., 1976; Luther, 1976).

Moreover, the $\Delta_1 T$ change from (33) has a negative feedback on $\Delta [O_3]$ (McElroy, et al. 1974). A positive δ gives an initial $+\phi$ in equation (29) and $-\Delta [O_3]$ and $-\Delta T$. Then k_1 and k_4 (destroying O_3) are decreased and k_5 (producing O_3) is increased. The second iteration of a perturbation thus tends to recover from the initial change. Blake and Lindzen (1973) point out that this effect also accelerates thermal relaxation compared with the rate, a , derived from infrared cooling alone.

Attenuation of sunlight affects J_7 directly; an effective attenuation cross section, analogous to equation (34), can be similarly defined and is numerically similar since the same radiation is involved. Once the first order $\Delta_1 T$ and $\Delta_1 J_7$ are

found for a given perturbation, δ , a revised equilibrium abundance, $[O_3]_2$ is computed with equations (8), (12), and (16) with these first-order corrections. Comparison with the first correction, $[O_3]_1$, gives the response of ozone to the effects of ΔT and ΔJ_7 alone. One can then readily estimate the convergent values, as follows:

Let the converged solution of $\Delta[O_3]$ be expressed as

$$\frac{\Delta[O_3]}{[O_3]} = \frac{\Delta_1[O_3]}{[O_3]} - \beta \frac{\Delta T}{T} = -\psi_1 \delta - \beta \frac{\Delta T}{T}, \quad (35)$$

where ψ_1 is the "initial" perturbation given by (29), β is a constant, and ΔT is the converged, final temperature change. From equation (33) the temperature change, $\Delta_1 T$, is nearly proportional to $\Delta_1 [O_3]$ and can be written generally as

$$\frac{\Delta T}{T} = \gamma \frac{\Delta[O_3]}{[O_3]}. \quad (36)$$

The first-order iteration is then

$$\frac{\Delta_1 T}{T} = \gamma \frac{\Delta_1 [O_3]}{[O_3]}, \quad (37)$$

which yields γ , since $\Delta_1 T$ and $\Delta_1 [O_3]$ are known. The equilibrium abundance, $[O_3]_2$, with the revised T and J_7 , gives

$$\frac{\Delta_2 [O_3] - \Delta_1 [O_3]}{[O_3]} = - \beta \frac{\Delta_1 T}{T}, \quad (38)$$

which yields β . Putting equation (36) into (35) we have the converged solution,

$$\frac{\Delta [O_3]}{[O_3]} = - \frac{\psi_1 \delta}{1 + \beta \gamma} \equiv - \psi \delta. \quad (39)$$

The temperature change may now be conveniently related to δ by a perturbation coefficient, τ , obtained from equations (36) and (39):

$$\frac{\Delta T}{T} = -(\gamma \psi) \delta \equiv -\tau \delta. \quad (40)$$

Finally an equilibrium calculation may be performed to test the convergence of the various perturbations that follow from a change, δ , in $[NO_x]$.

Another feedback effect noted by McElroy et al. (1974) is the level of production of $O(^1\Delta)$; if O_3 is reduced, the natural production of NO_x from $N_2O + O(^1\Delta)$ is increased at low altitudes and decreased at higher ones. The overall effect

is small and can be ignored anyway if we regard δ as the net change in $[\text{NO}_x]$.

The input data and the results of this analysis are explained in Figures 1 and 2. A positive ψ and τ mean that both $[\text{O}_3]$ and T decrease as $[\text{NO}_x]$ increases. Unfortunately with the present approach the temperature decrease cannot be followed all the way to the tropopause, because at large optical thicknesses the concept of a local cooling rate balancing the local heating by sunlight no longer governs the temperature. Radiative exchange among different parts of the atmosphere and the ground becomes dominant. The principal effect of decreasing O_3 in the lower stratosphere is to decrease the temperature because less radiation from the ground is absorbed by O_3 in the $9.6\mu\text{m}$ region (Ramanathan, et al., 1976).

3. Discussion: The Effect of Stratospheric Ozone on Climate

The usual (but not unanimous) conclusion from studies with radiative-convective models (e.g., Manabe and Wetherald, 1967; Ramanathan, et al., 1976) is that lowered stratospheric ozone will lower surface temperatures. Nevertheless, the feedback processes are numerous and complex. One of the more important aspects of ozone is its dynamical transport and relatively inert chemistry in the lower stratosphere. Dickinson (1974) has cautioned that we know very little about how climatic changes would feed back

on stratospheric dynamics, which must be coupled to circulation in the troposphere. Evidently an increased surface heating by low lying absorbing particles could even reverse (Reck, 1976) the net cooling calculated for no absorbing particles and an ozone depletion (e.g., Ramanathan, et al., 1976).

Thus one way in which reductions in ozone could initiate colder climatic periods is through radiative-convective impact of the stratosphere on the surface. It appears that the increased solar flux reaching the surface, with an ozone reduction, is more than compensated by surface cooling through reduction of the greenhouse effect at the $9.6 \mu\text{m}$ vibration - rotation band (Ramanathan et al., 1976).

A second, and primarily stratospheric, possibility which seems to deserve rather more attention is that a decrease in ozone will lead to haze or cloud formation in the lower stratosphere, significantly raising the Earth's albedo. There are several items that seem to bear on this possibility.

(1) The H_2O mixing level in the lower stratosphere (around 5×10^{-6} by volume) is close to the saturation vapor pressure at the tropical tropopause ($\sim 205^\circ\text{K}$ at 16 km). On the pressure scale the H_2O is a factor of about 5 too low for condensation to occur; but on the temperature scale the tropic tropopause averages only about 10°K above the

saturation point. This coincidence has led to the common belief (Harteck and Jensen, 1948; Mastenbrook, 1971) that the "cold trap" of the tropopause sets the upper limit to the mixing ratio there and above. Even under present conditions stratospheric clouds are not unknown. Newell (1972) believes that the stratospheric mother-of-pearl clouds occasionally seen over Iceland in the winter arise from moist air systematically forced upward in its zonal flow.

(2) Not only does a decrease in ozone decrease the stratospheric temperature, but the temperature profile near the tropopause is altered. Manabe and Strickler (1964) and Reck (1976) find the tropopause to be erased by a complete (presumably catastrophic) removal of O_3 , independent of water clouds or aerosols in the models. A change in the dynamical interaction of the troposphere and stratosphere could be most important to the moisture content and condensation in the stratosphere.

(3) Condensation nuclei are, of course, important. It would be useful to understand how aerosol chemistry would change in the stratosphere with an ozone reduction. In addition, if the O_3 reduction is related to increased cosmic-ray ionization reaching the lower stratosphere, as proposed in this paper, the temperature decrease in conjunction with an increased ionization rate could be

responsible for increasing the albedo. The ion densities in the stratosphere are the order of 10^4 cm^{-3} (Ruderman, et al., 1976), with both positive and negative ions being hydrated with several water molecules. Presumably larger initial water clusters are needed for continued growth. The role of these ions in attaching to aerosols before they are electrically neutralized (with lifetimes of about 300 sec) is not well understood.

(4) Long-term feedbacks involving the oceans, ice-caps, and circulation of the troposphere are not easy to predict, and natural oscillations within the system are conceivable. But there is a first-order feedback that acts in the positive or reinforcing direction and is likely to be exceptionally important. A small decrease in stratospheric ozone probably decreases the ground temperature, whether by condensation of stratospheric moisture or merely by opening the $9.6\mu\text{m}$ cooling window. The cooler ground radiates less energy to the region just above the tropopause, which is heated through infrared absorption by ozone (as well as CO_2 and H_2O) and not so much by ultraviolet absorption of sunlight. This is the crucial altitude for increased condensation, and the diminished tropopause temperature will lead to further condensation. And so on.

One problem, however, that does not seem to be settled is whether condensation of stratospheric water would cause enough heating by infrared absorption to offset the increased albedo to sunlight. The formation of high cirrus

clouds in the troposphere probably does heat not only the tropopause but the ground (Manabe and Strickler, 1964). But it is not clear to what extent condensation in a nearly saturated stratosphere would modify the warming effects there compared with the warming caused by the low (absolute) humidity already present.

I am grateful to Steven Wofsy and Michael McElroy for supplying me with the detailed data that were used in developing their 1974 model of the stratosphere, which served as the starting point for the perturbation analysis of section 2. The research reported in this paper was supported by the Atmospheric Sciences Section of the National Science Foundation and the National Aeronautics and Space Administration.

Legends

Fig. 1. Adopted height distribution of odd nitrogen and oxygen in the unperturbed atmosphere. The model is based on that of McElroy et al. (1974), which was modified to give self-consistent solutions for a closed system of reactions (1) through (7) in photochemical equilibrium above 35 km. Reaction rates used are those quoted by McElroy et al., who also supplied photolysis and heating rates, which were altered to fit the modified model.

Fig. 2. Perturbation coefficients for $\Delta[O_3]$ and ΔT . For ψ the dashed line (ψ_1) is defined by equation (29); the solid line is ψ given by equation (39). For τ the dashed line is $\tau_1 = -\Delta_1 T / T_0$, where $\Delta_1 T$ is given by equation (33); the solid line is $\tau = \tau_1 \psi$ from equation (40). The ψ_1 and τ_1 are independent of δ except for attenuation of sunlight by perturbations overhead. The final ψ and τ do depend on δ which was taken uniformly through the atmosphere as $\delta = +0.1$. Below 35 km the τ 's computed by this technique are unreliable, because Dickinson's (1973) linear perturbation treatment of radiative cooling is not appropriate. Below 35 km mixing is more rapid than photochemical relaxation and we take the ψ perturbation coefficients as constant and fixed by the 35-km values.

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